

# The Effect of Stainless-Steel Containers on High-Resolution Gamma-Ray Analysis of Plutonium

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This article was submitted to  
5<sup>th</sup> International Conference on Methods and Applications of  
Radioanalytical Chemistry-Marc V  
Kailua-Kona, HA  
April 9-14, 2000

**U.S. Department of Energy**

Lawrence  
Livermore  
National  
Laboratory

**March 21, 2000**

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# THE EFFECT OF STAINLESS-STEEL CONTAINERS ON HIGH-RESOLUTION GAMMA-RAY ANALYSIS OF PLUTONIUM

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## EXECUTIVE SUMMARY

The 240-Pu / 239-Pu ratio and age of plutonium, contained within a 0.25 inch thick stainless-steel can, can be quickly and accurately determined using a high-resolution gamma-ray detector and the gamma-ray analysis program MGA. Counting times as low as 3 minutes will determine the 240-Pu / 240-Pu ratio to within 5%. Plutonium with containers as thick as 0.5 inches can be accurately analyzed for both 240-Pu / 239-Pu and plutonium age.

## ABSTRACT

The goal of this work was to determine the effects on plutonium isotopic analysis of having plutonium inside of a 0.25 inch thick stainless steel can. To do this, we analyzed plutonium samples with a U-Pu InSpector (which uses a high-resolution gamma-ray detector and the analysis code MGA (Multi Group Analysis)), to determine both the 240-Pu / 239-Pu ratio and the years since the plutonium was separated from americium. We analyzed a 1.6 kg plutonium sample that was placed inside of a 0.25 inch can at varying distances (0-2 meters) and count times (10 seconds – 30 minutes). In separate experiments, we analyzed 0.4g plutonium sources with stainless-steel thickness' ranging from 0.125 to 1.0 inch. This report will show three effects of having plutonium in a stainless steel can

1. 240-Pu / 240-Pu can be quickly and accurately determined for a 1.6 kg plutonium sample inside of a 0.25 inch thick stainless-steel can, as this thickness of stainless steel acts as a perfect filter to reduce the intense 59keV gamma peak from 241-Am.
2. The accuracy of determining the plutonium-americium separation date is not effected by 0.25 inch of stainless steel.
3. Both 240-Pu / 239-Pu and the americium separation date can be accurately determined for stainless-steel thickness' up to 0.5 inches, requiring additional counting time with increasing thickness to obtain desired precision. For stainless-steel absorber thickness' greater than 0.5 inch, MGA analyses are not reliable.

## EXPERIMENT ONE: COUNTS WITH A 1.6 KG PLUTONIUM OXIDE SAMPLE INSIDE OF A \_ INCH THICK STAINLESS-STEEL CAN

Two preliminary background counts (one close to the stainless-steel can, one at 1 meter) were done to insure that there were no interfering gamma peaks. No plutonium or americium peaks were seen in either count.

The 240-Pu / 239-Pu ratio and americium separation date of the plutonium oxide were previously determined (using MGA in a 1995 plutonium inventory analysis). We re-measured the oxide (without the 0.25 inch stainless steel can) to confirm these values. For optimal analysis with MGA, the intense gamma peak at 59keV from 241-Am must be reduced, so that the ratio of the Am peak to the most intense plutonium peak in the 100keV region is between 0.1 and 10. With the U-Pu InSpector, adding one or two tin filters usually does this. We counted the plutonium twice: once using one tin filter, and once using two. The weighted average of the two measured 240-Pu / 239-Pu values,  $.0634 \pm 0.8\%$ , agreed with the 1995 value ( $0.0622 \pm .6\%$ ) within two sigma. The 1995 value was used to determine the accuracy of the current 240-Pu / 239-Pu determinations with the plutonium in the can. The re-measured americium separation dates also agreed with the 1995 data, when corrected for the difference in measurement dates. The time corrected 1995 americium separation date was used for comparison to the current plutonium counts.

A total of 26 measurements were done with the plutonium inside the 0.25 inch thick stainless-steel can. For these measurements, no tin filters were added, as the 0.25 inch of stainless steel reduced the Am peak to the same height as the plutonium peaks. All counts were done with the inspector "aimed" at the side of the can, at the center of the plutonium oxide. Count times ranged from 10 seconds to 30 minutes, and at six distances between 0.004 meters (touching the stainless-steel can) to 2.0 meters. We used the collimator to keep the dead time below the recommended maximum of 20% for counts closer than 0.242 meters. Measurements both with and without the collimator were taken at .242 meters.

#### EXPERIMENT ONE RESULTS: 240-Pu / 239-Pu

All counts except one resulted in a MGA report. The exception (a 15-second count at .242 meters with the collimator)) did not have enough counts for MGA to analyze. Figure 1 shows the 240-Pu / 239-Pu values reported by MGA. All 25 data points are displayed, with their one-sigma error bars. The data is plotted to compare with the "true" 240-Pu / 239-Pu value of .0622, as shown with the middle horizontal line. The count times at each distance are shown: count times listed above the center line were done without the collimator, while times below the center line were done with the collimator in place.

The ratio of the reported values with the true 240-Pu / 239-Pu is shown in figure 2. In this graph, the distance value of the data points were offset to allow viewing of all data points and error bars. The data was offset so that the longest count is shown to the right. For example, the three data points centered around 1.5 meters are for 2, 5, and 10-minute counts, going left to right. The data agrees well with the expected 240-Pu / 239-Pu value, as the number of data points that agree within their one sigma error is 20 out of 25, within the range expected ( $17 \pm 4$ ). The (unweighted) average of all 25 ratios is 0.999

Figure 3 shows a representative time slice of the data, graphing results from 15-minute counts (with no collimator) at different distances. The linearly increasing error with increasing distance is shown in figure 4.

Figure 5 graphs data taken at the maximum count rate, at .242 meters, for count times from 15 seconds to 15 minutes. The decrease in the reported 240-Pu / 239-Pu one-sigma error with increasing count time is shown in figure 6, and the linear fit of the error bar data to  $1/\text{square root}(\text{time})$  is shown in figure 7. From this graph, we plotted (figure 8) the count time needed at .242 meters to obtain a 240-Pu / 239-Pu ratio with a desired degree of accuracy, with 95% certainty.

In a similar manner, we determined the time needed to obtain a desired precision at distances greater than .242 meters, as shown in figure 9.

Further experimental details are given in appendix A

#### EXPERIMENT ONE RESULTS: YEARS SINCE CHEMICAL SEPARATION FROM AMERICIUM

MGA also reported the years since the separation of the plutonium from americium, with the associated error. This data is plotted in figure 10. The ratio of the reported value to the true value is shown in figure 11. Note that the reported value agrees with the true value for all but two counts.

#### EXPERIMENT TWO: EFFECT OF DIFFERENT THICKNESSS OF STAINLESS- STEEL CANS

We used the same U-Pu InSpector to determine the effect of different thickness' of stainless-steel cans on the 240-Pu / 239-Pu ratio and the americium separation date. Thicknesses between .125 and 1.0 inch of stainless steel (in 1/8 inch increments) were placed between the source and the detector. The plutonium sample used (pidie #1, 0.4g plutonium oxide) had a 240-Pu / 239-Pu ratio of .0636. The distance between the source and the detector was kept constant (1 inch), as was the count time (30 minutes), for measurements with thickness' from .125 to .875 inches. The measurement with 1 inch of stainless steel was counted for 2400 minutes. No tin filters were used in any of the counts

#### EXPERIMENT TWO RESULTS.

Both 240-Pu / 239-Pu and the americium separation date were accurately determined for stainless-steel thickness' up to 0.5 inch. Figure 12 shows the effect of stainless-steel thickness on 240-Pu / 239-Pu. Values for thickness of .5 inch and below were within 2 sigma of the true ratio. The increase of the reported 240-Pu / 239-Pu error with increased stainless-steel thickness is shown in figure 13. Figure 14 shows the effect on the reported years since americium separation, and figure 15 the reported error on the separation interval.

Results were not reliable for thickness' greater than .5 inch. This was seen in different ways for these counts, including:

1. Some measured values of 240-Pu / 239-Pu did not agree with the true value
2. Some incorrect plutonium ages were reported
3. MGA reported warning messages, related to the quality of the analysis, for all counts with more than 0.5 inch of stainless-steel absorber.
4. The reported value of NQFIT exceeded 1.05 on some counts, indicating a poor fit to the spectrum

5. The reported cadmium thickness became non-linear above 0.625 inch

### EXPERIMENT THREE: STATISTICAL PRECISION OF REPORTED ERRORS OF (240-Pu / 239-Pu) AND AMERICIUM SEPARATION DATES

We did a series of 40 counts on a 0.4g plutonium sample (Pidie #3), to demonstrate the statistical validity of the reported one-sigma error of the 240-Pu / 239-Pu, and the reported error of the plutonium age. Each count was for 10 minutes. There was no data available on the true date of americium separation for this source.

### EXPERIMENT THREE RESULTS

The 240-Pu / 239-Pu results are shown in figure 16. The data is consistent with the expected distribution, as 27 of 40 are within one sigma (expect  $27 \pm 5$ ), 11 between one and two sigma (expect  $11 \pm 3$ ) and 2 are greater than two sigma (expect  $2 \pm 2$ ).

The reported error of the americium separation date did not show such agreement, however. All 40 reported values agreed within their error range with the average value (figure 17).

### CONCLUSIONS

We used a U-Pu InSpector, with the data analysis code MGA, to determine the effects on plutonium isotopic analysis of having a 1.6kg plutonium oxide sample within a 0.25 inch thick can. We conclude that the same accuracy of the (239-Pu / 240-Pu) ratio and americium separation date is obtained with the 0.25 inch stainless-steel can as without it. We also conclude that the reported one-sigma error for the 240-Pu / 239-Pu follows the expected statistical distributions, and that the reported americium separation date is correct within its reported error at least 95% of the time.

We determined the necessary count length to obtain a desired precision in the reported 240-Pu / 239-Pu, with a 95% confidence level. For counts at the maximum recommended count rate, these range from 67 minutes for 1% precision to less than one minute at 10% precision.

We also conclude that plutonium can be successfully analyzed with the U-Pu InSpector through a maximum of 0.5 inches of stainless steel. Beyond that, the reported results are not reliable.

## APPENDIX A. EXPERIMENTAL DETAILS

**LOCATION:** The counting of the 1.6kg plutonium sample was done in building 332, room 1346. The sample and detector were placed on adjustable height tables so that the center of the detector and the center of the plutonium oxide were both at a height of 100.5 cm. The sample was located 1.7 meters from the wall behind it. To measure at different sample-detector distances, the detector was moved, while the source remained stationary.

**DETECTOR:** We used a Canberra U-Pu InSpector, which includes a high-resolution germanium gamma-ray detector, a collimator, and three tin filters. The detector has a surface area of 5 cm<sup>2</sup>, has a maximum count rate of 50,000 cps and a FWHM of 550-600eV @ 122keV. The collimator has a 0.8 cm<sup>2</sup> opening. The three tin filters are each 0.8 mm thick.

**ANALYSIS SOFTWARE: MGA.** We used the Canberra version of the MGA (multi-group analysis) software to analyze the plutonium gamma spectra. The code uses gamma peaks in the 100keV region to determine plutonium isotopes. In addition to 240-Pu / 2390-Pu (and its associated one sigma error), MGA reported

- Dead time
- Quality of analysis parameters (QFIT and NQFIT)
- Other plutonium isotopes (238-Pu, 241-Pu) and their one sigma errors
- Estimated 242-Pu
- 241-Am and its one sigma error
- Full width at half maximum (FWHM) at 122keV and 208keV
- Total counts
- Plutonium thickness (Pu g/cm<sup>2</sup>)
- Filter Thickness (Cd g/cm<sup>2</sup>)
- Years since Americium separation, with error

Data was taken both with and without the collimator. The FWHM @ 122keV for all counts were in the specified range. The maximum count rate in these experiments was 5,533 cps, well below the maximum.

All counts were done with dead time below 25%. All reports from the first experiment had NQFIT values below 1.05, indicating good fits to the data were obtained. Total counts ranged from 30,000 – 7,460,000.

**PU SAMPLES:** We used a single sample of plutonium oxide to determine the effects of 0.25 inch stainless steel on the 240-Pu / 239-Pu analysis and the americium separation date. The 240-Pu / 239-Pu ratio of the plutonium oxide was previously determined (using MGA in a 1995 plutonium inventory analysis) to be  $0.0622 \pm .6\%$ . The sample had 1.6 kg of Plutonium and was doubly contained in two cans, each with .254 inch thick walls. The inner can had an inner diameter of 106 mm, and was filled to a height of 9.5 cm with plutonium oxide. The can was shaken to level the Pu oxide before the counts.

We used a 0.4g Pidie plutonium standard to measure the effects of varying the stainless-steel thickness. This source had a 240-Pu / 239-Pu ratio of .0636.

**STAINLESS-STEEL ABSORBERS:** The 1.6kg Pu sample was contained within two stainless-steel cans, each with a thickness of .01 inch. Measurements of the 1.6kg Plutonium sample were done with the sample placed inside of an additional can of stainless steel. The can consisted of two pieces: an inner can, 0.125 inches thick, and an outer liner, also 0.125 inches thick. The total thickness was .27 inches of stainless steel.

In the second experiments, from one to eight stainless-steel plates (each 0.125 inch thick.) were used as absorbers. The plutonium source was in a stainless-steel container, .01 inch thick at the measured surface.

This work was performed under the auspices of the U.S. Department of Energy by University of California  
Lawrence Livermore National Laboratory under contract No. W-7405-ENG-48.

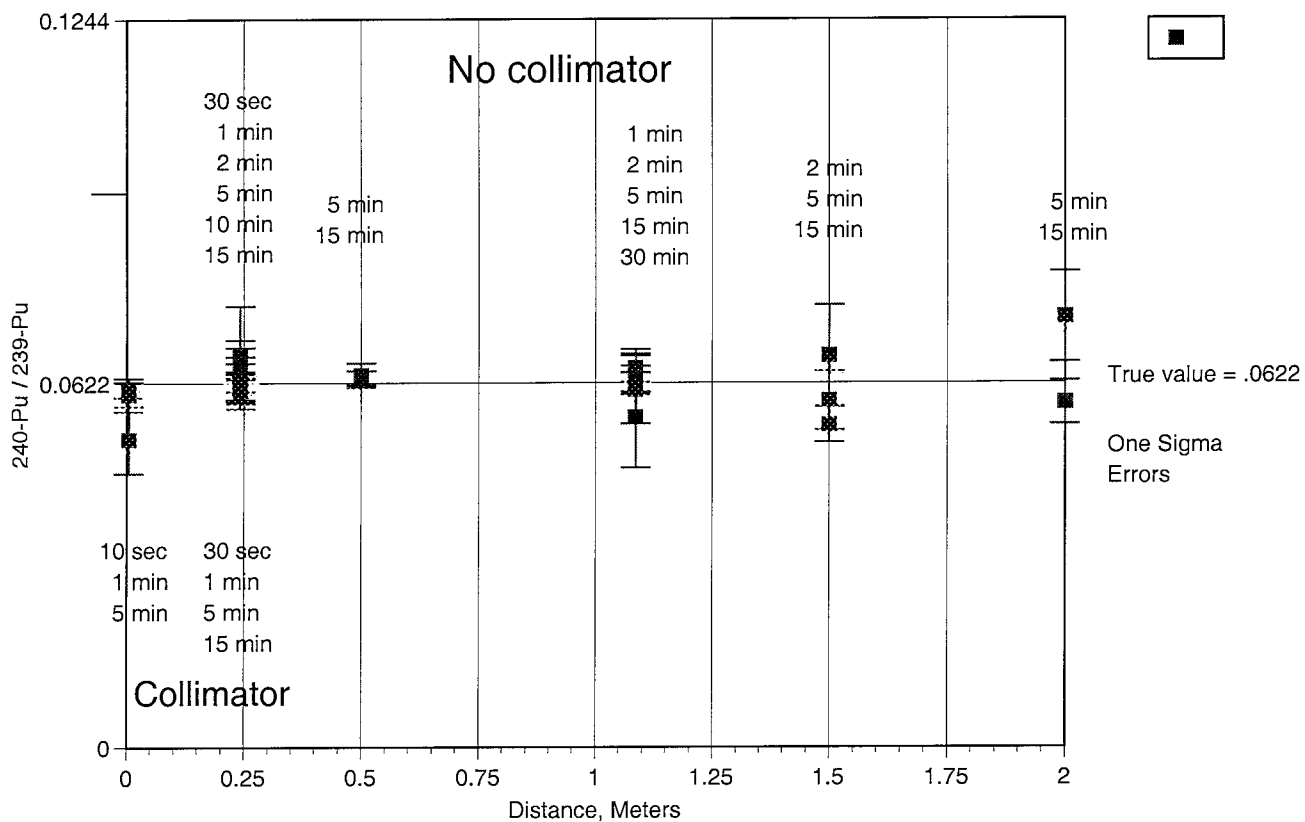


Figure 1.  $^{240}\text{Pu} / ^{239}\text{Pu}$  was determined between 0 and 2 meters, with count times from 10 seconds to 30 minutes

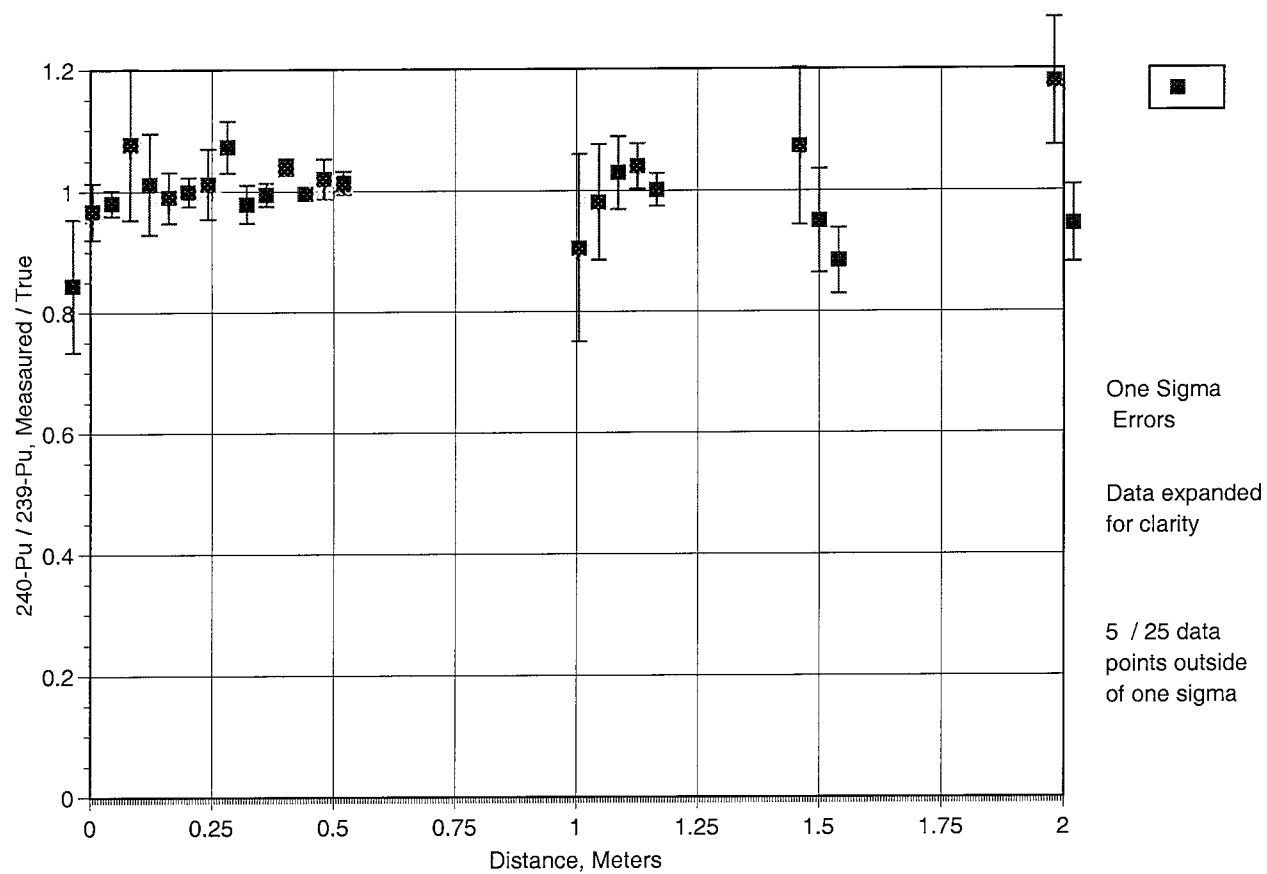


Figure 2.  $^{240}\text{Pu} / ^{239}\text{Pu}$  was accurately determined (expanded view)

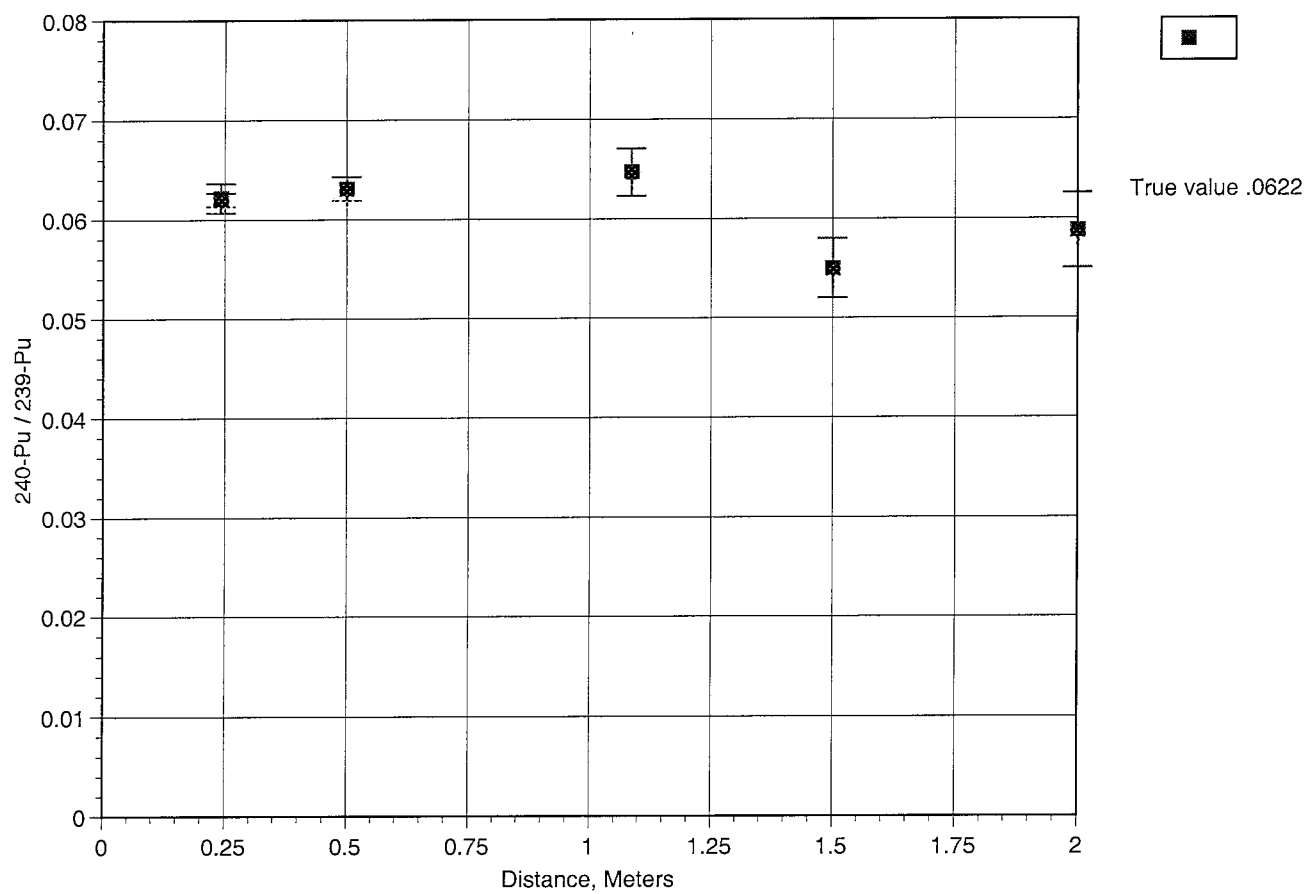


Figure 3.  $^{240}\text{Pu} / ^{239}\text{Pu}$ , 15 minute counts

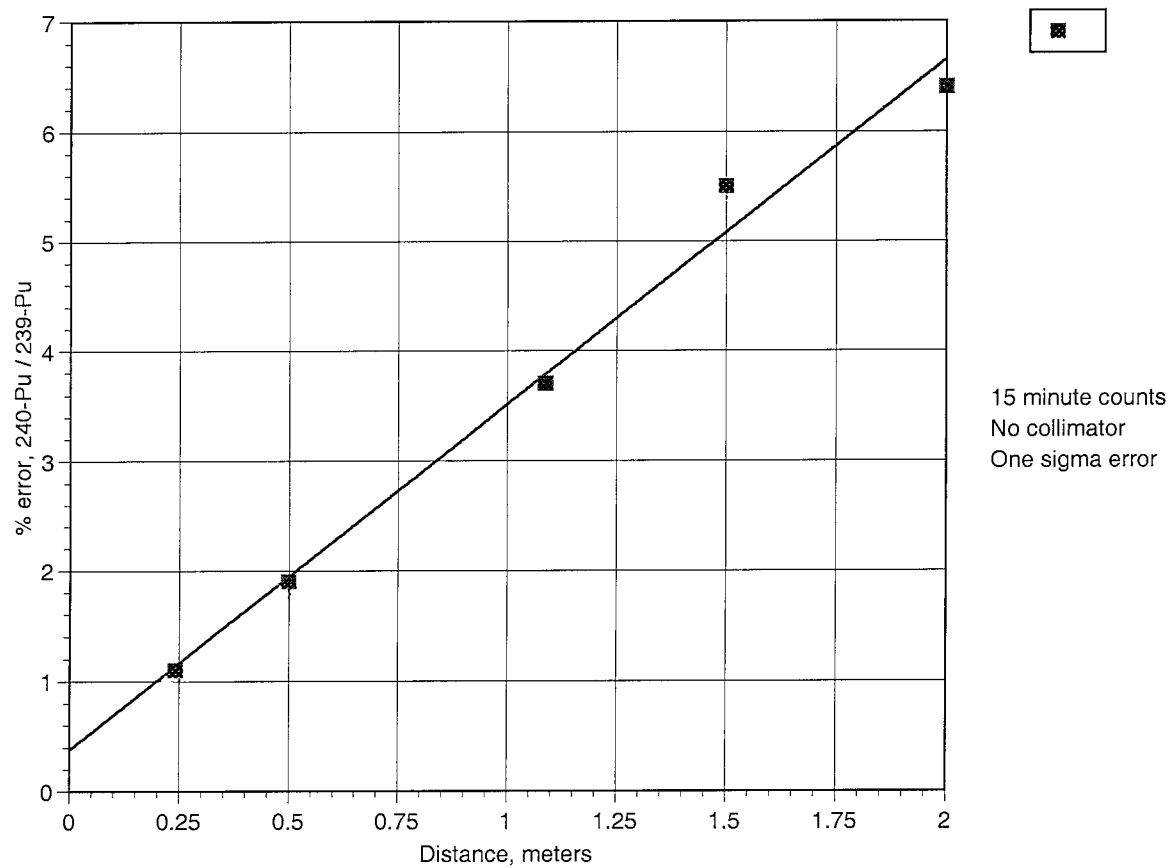


Figure 4. % error in  $^{240}\text{Pu} / ^{239}\text{Pu}$  is proportional to the distance

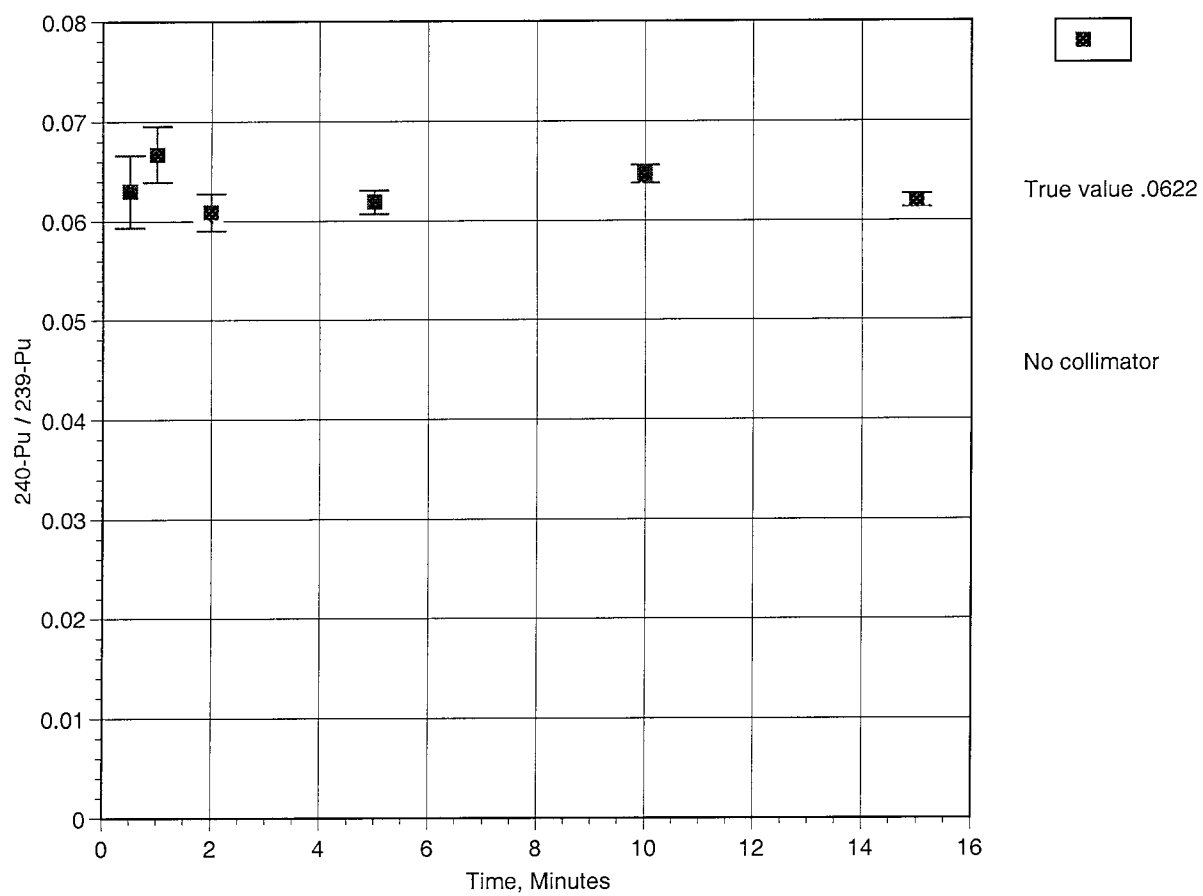


Figure 5.  $^{240}\text{Pu} / ^{239}\text{Pu}$  at .242 Meters

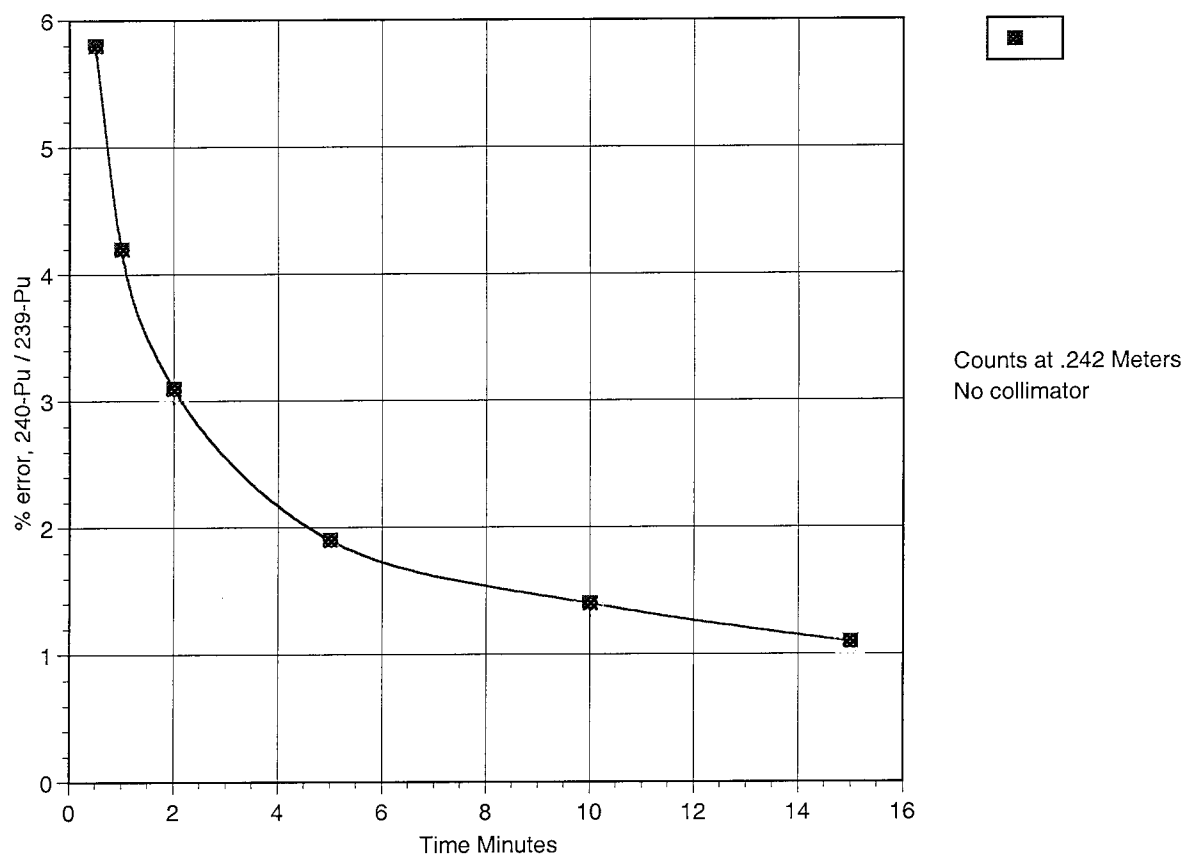


Figure 6. % error (240-Pu / 239-Pu ) decreases with increasing count time

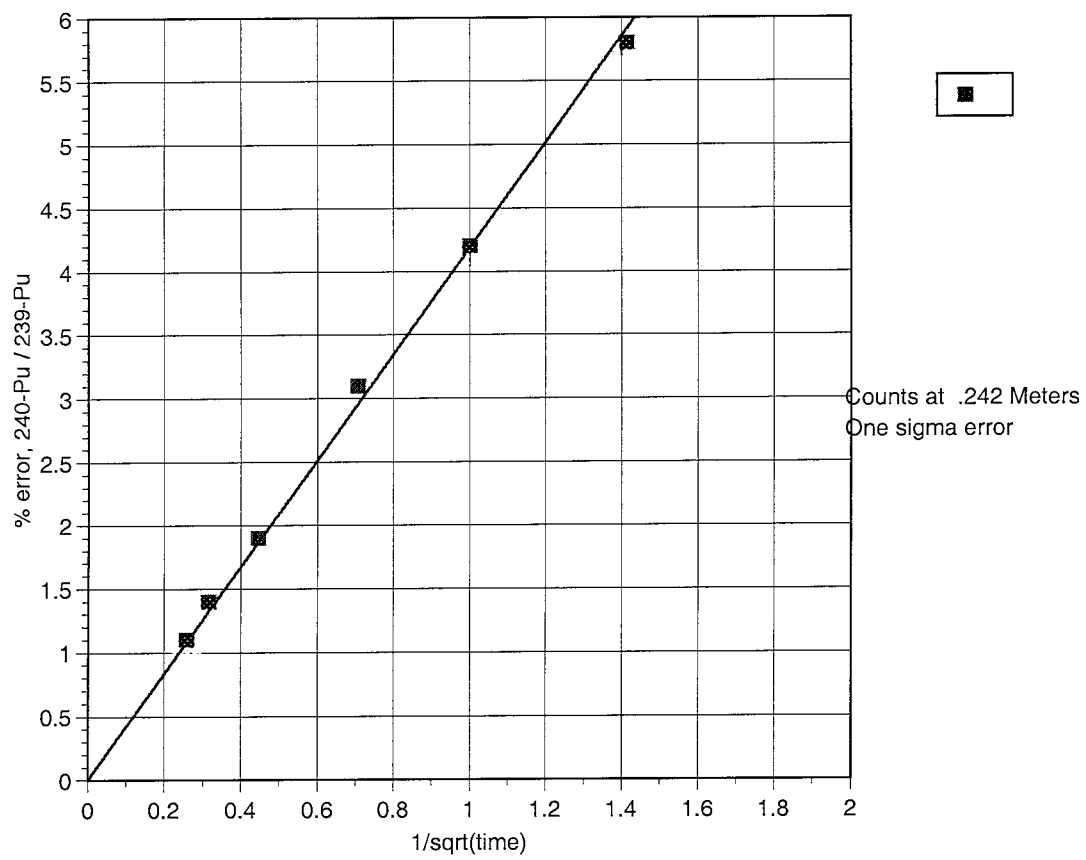


Figure 7. % error (240-Pu / 239-Pu) is proportional to  $1/\sqrt{\text{time}}$

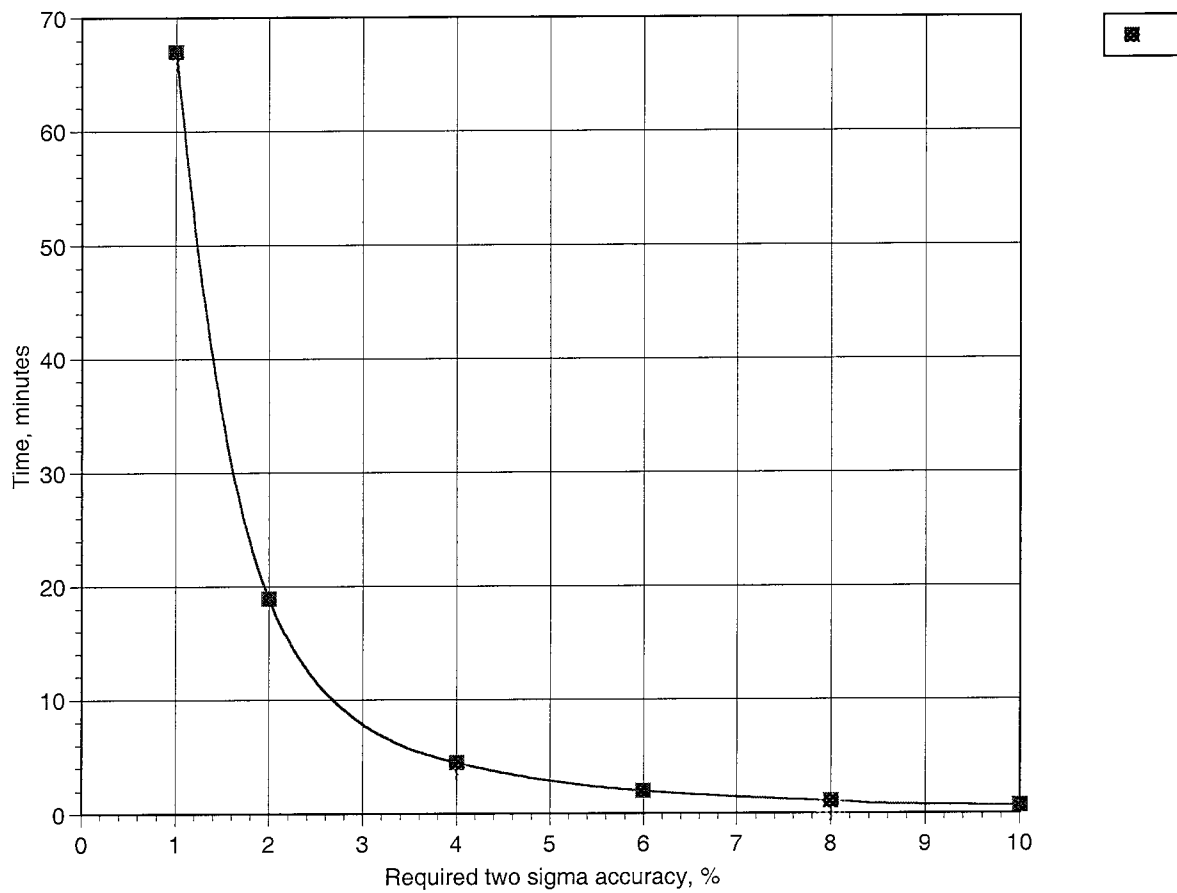


Figure 8. How long do I need to count to get a required 240-Pu / 239-Pu precision?

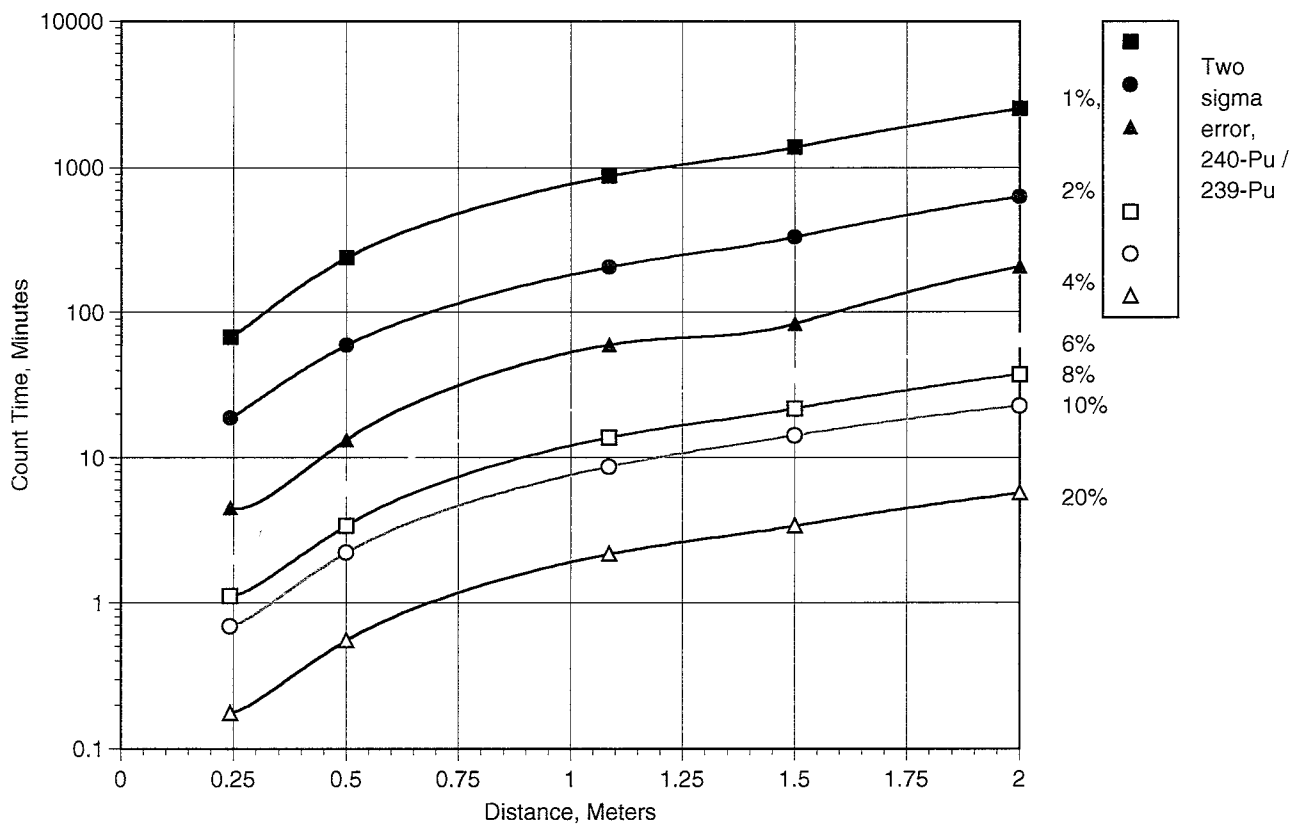


Figure 9. How long do I need to count to determine 240-Pu / 239-Pu with 95% certainty?

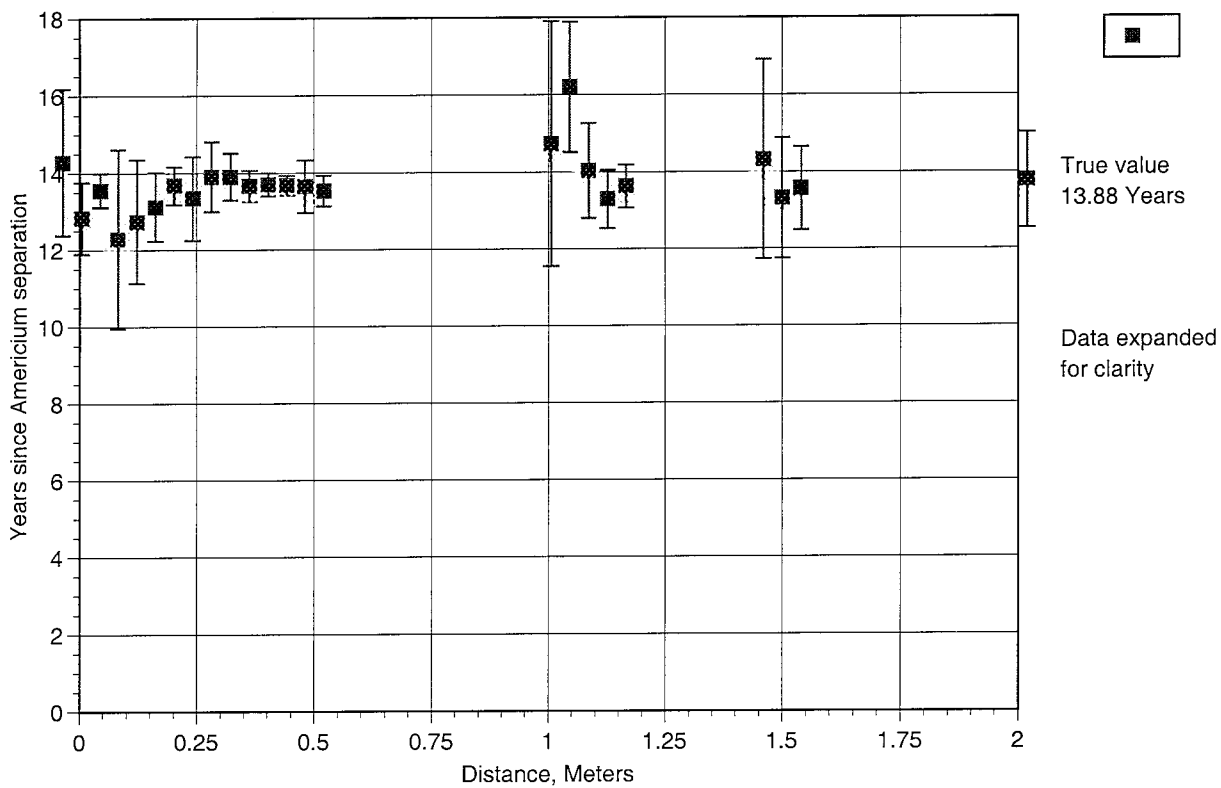


Figure 10. The time since chemical separation of Americium was determined

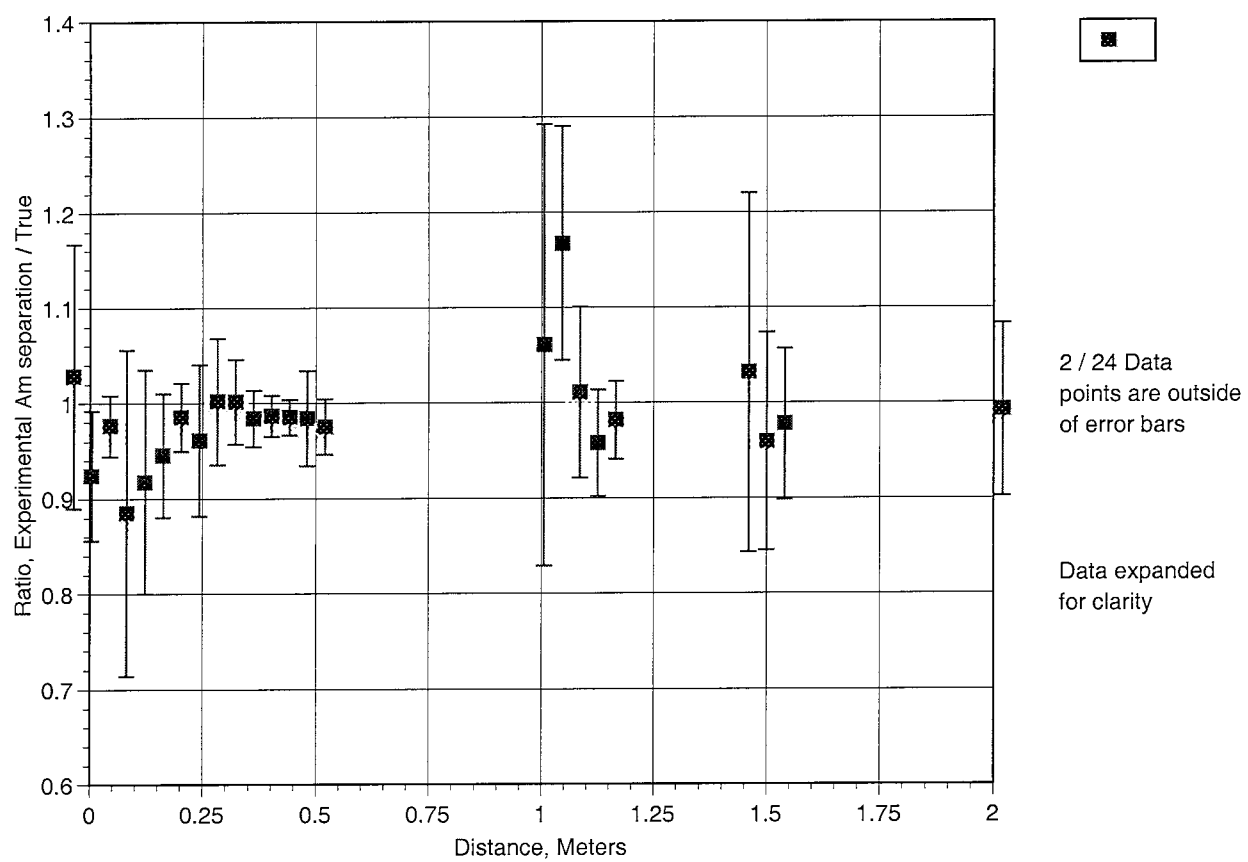


Figure 11. The time since Americium separation was correctly determined at all distances

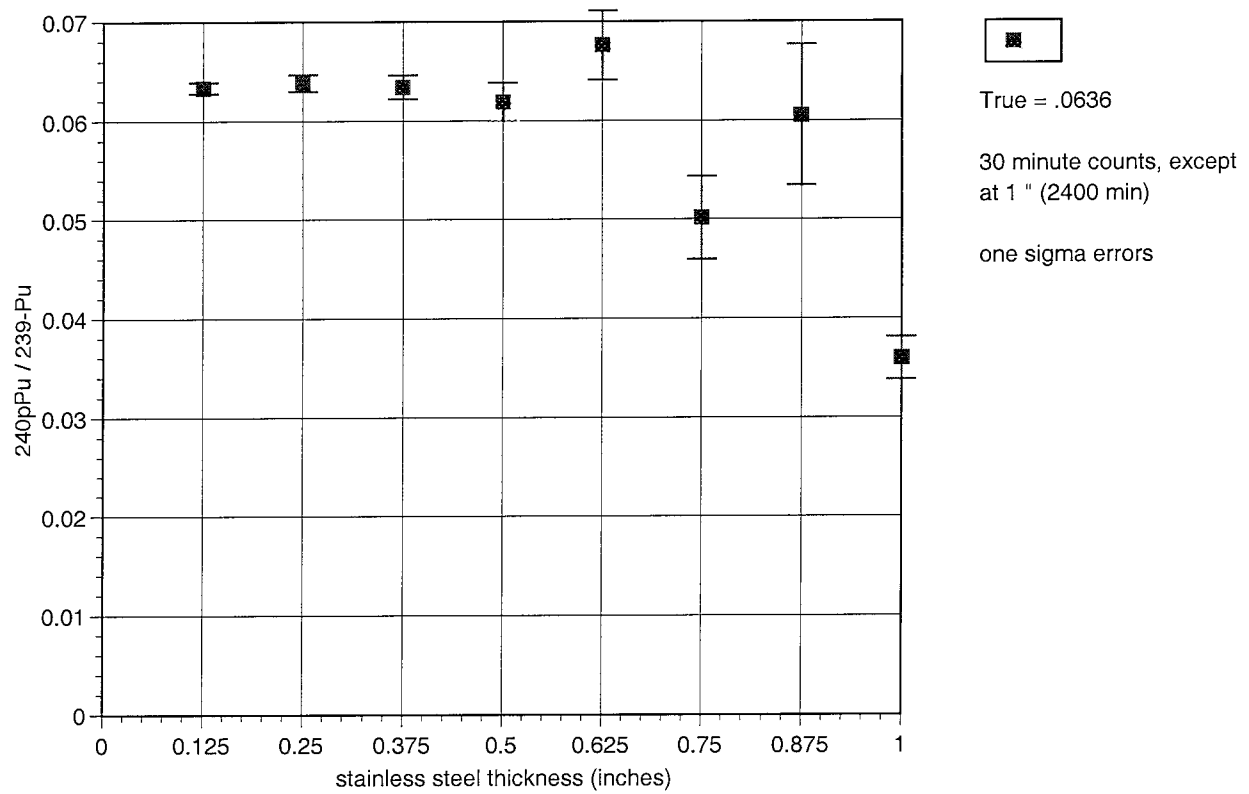


Figure 12.  $^{240}\text{Pu} / ^{239}\text{Pu}$  was accurately determined for stainless steel thicknesses up to 0.5"

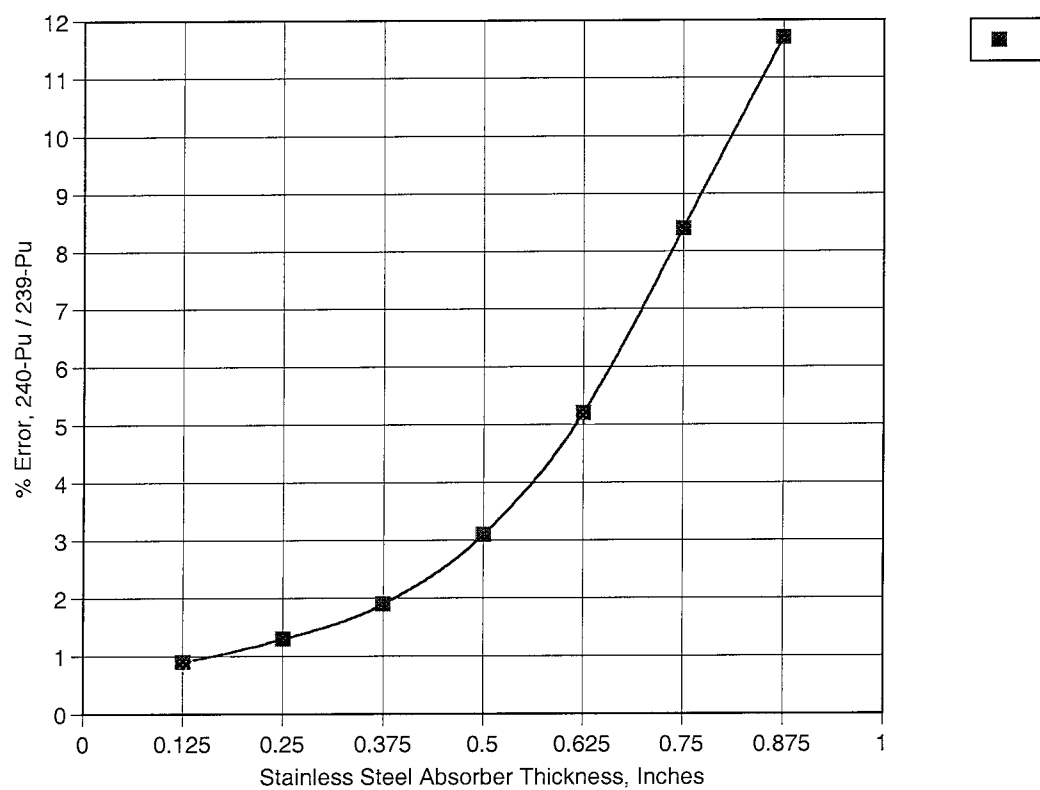


Figure 13. 240-Pu / 239-Pu error increases with increasing absorber thickness

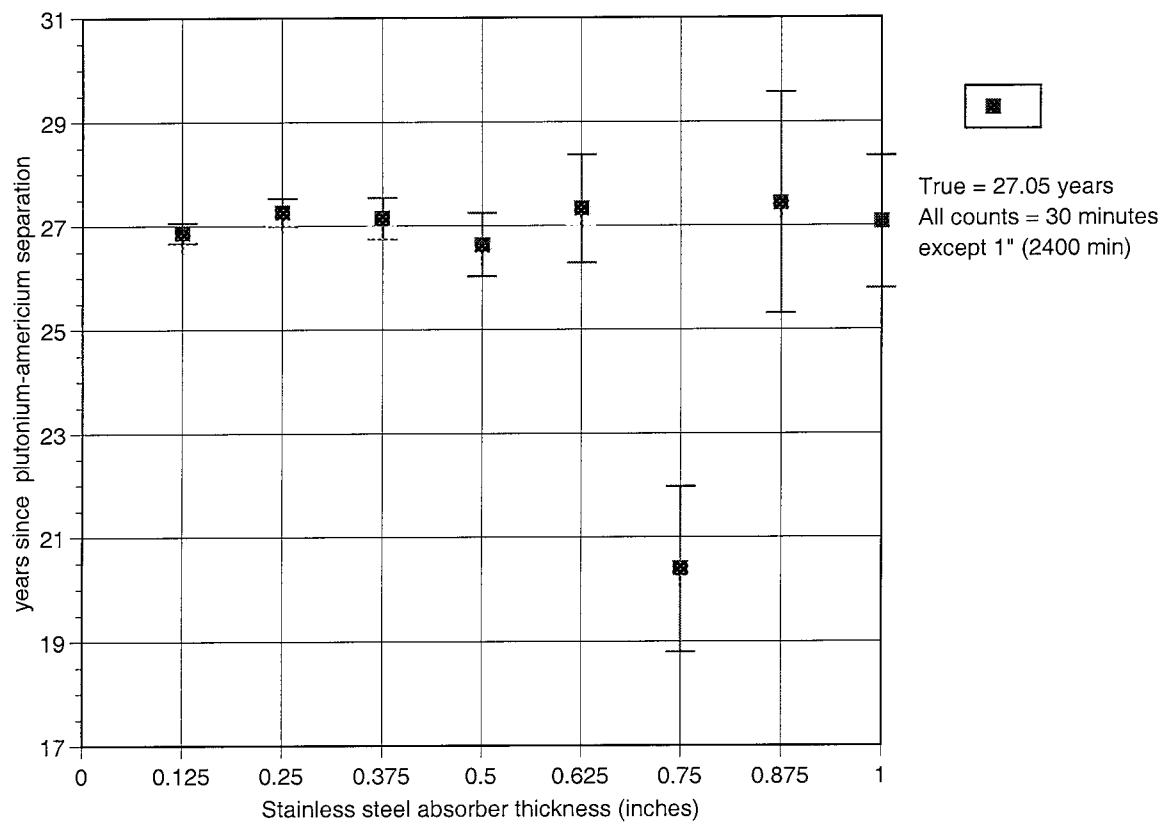


Figure 14. Reported Am separation date is correct for stainless steel thicknesses less than 0.5 inches

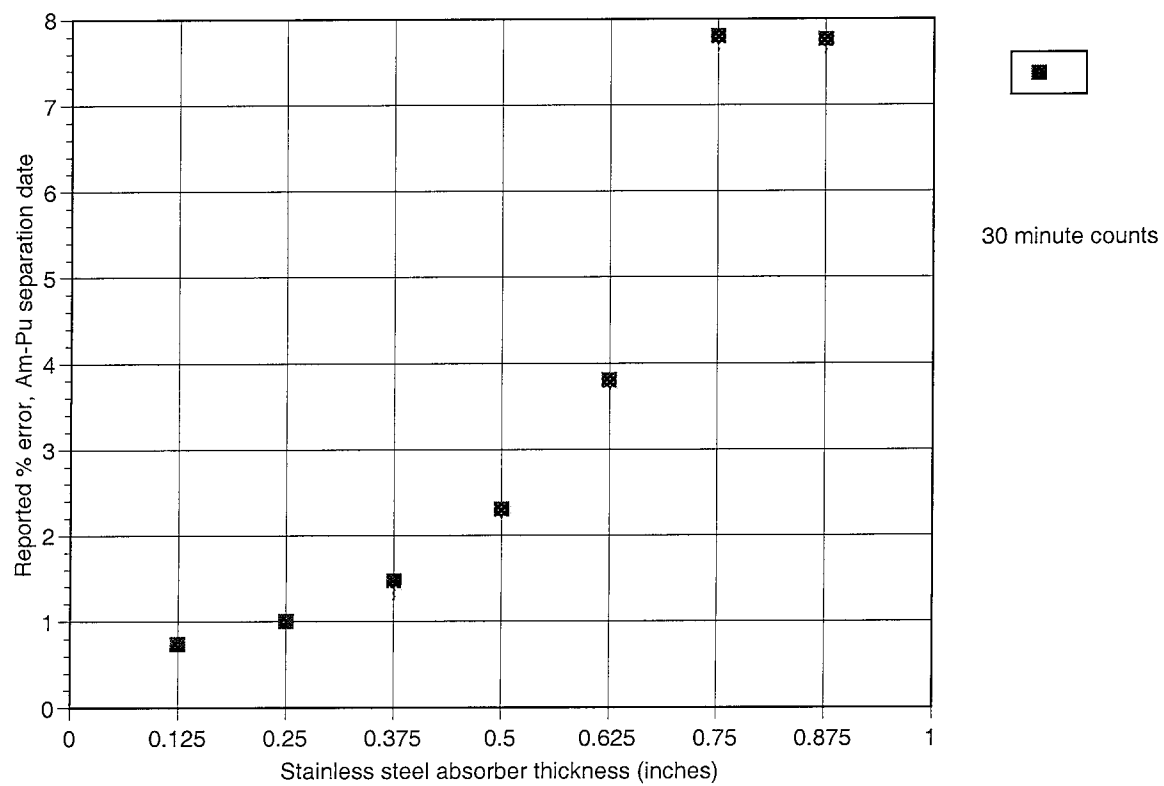


Figure 15. Americium-plutonium separation date error increases with stainless-steel absorber thickness

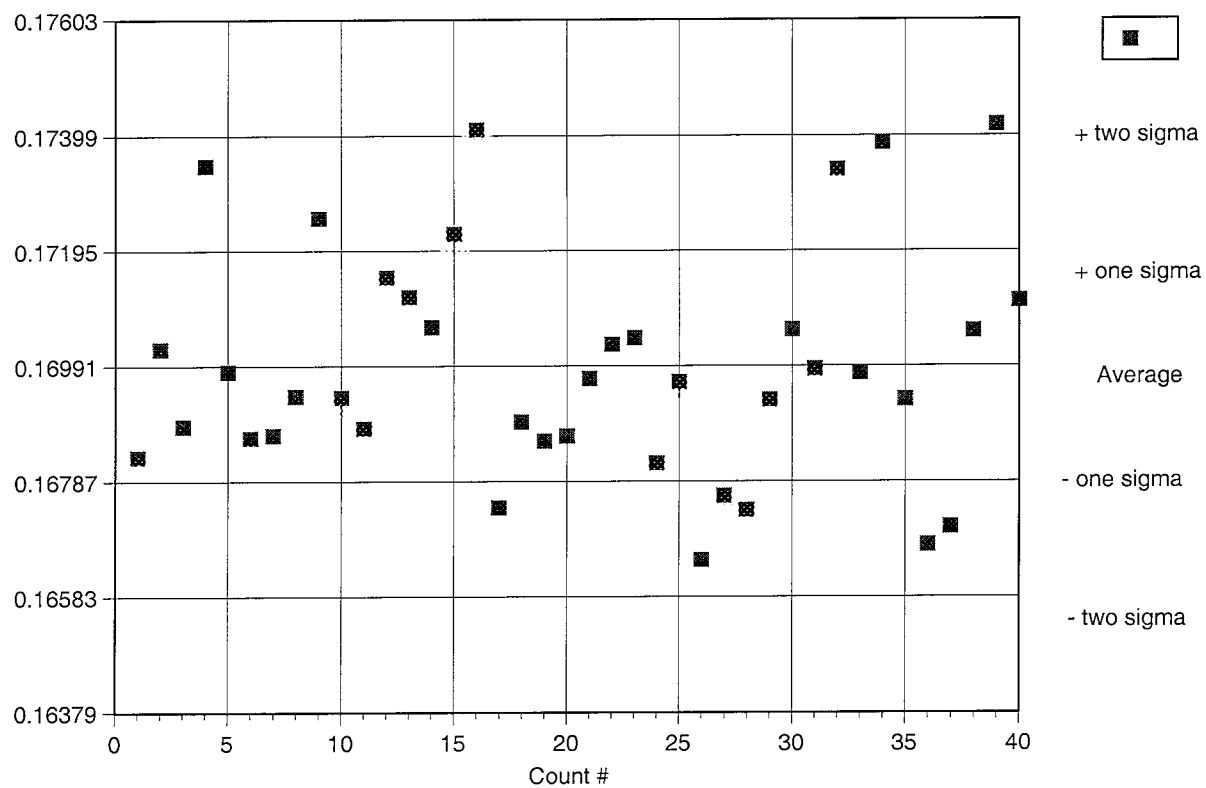


Figure 16.  $^{240}\text{Pu}$  /  $^{239}\text{Pu}$  precision agrees with reported one sigma errors

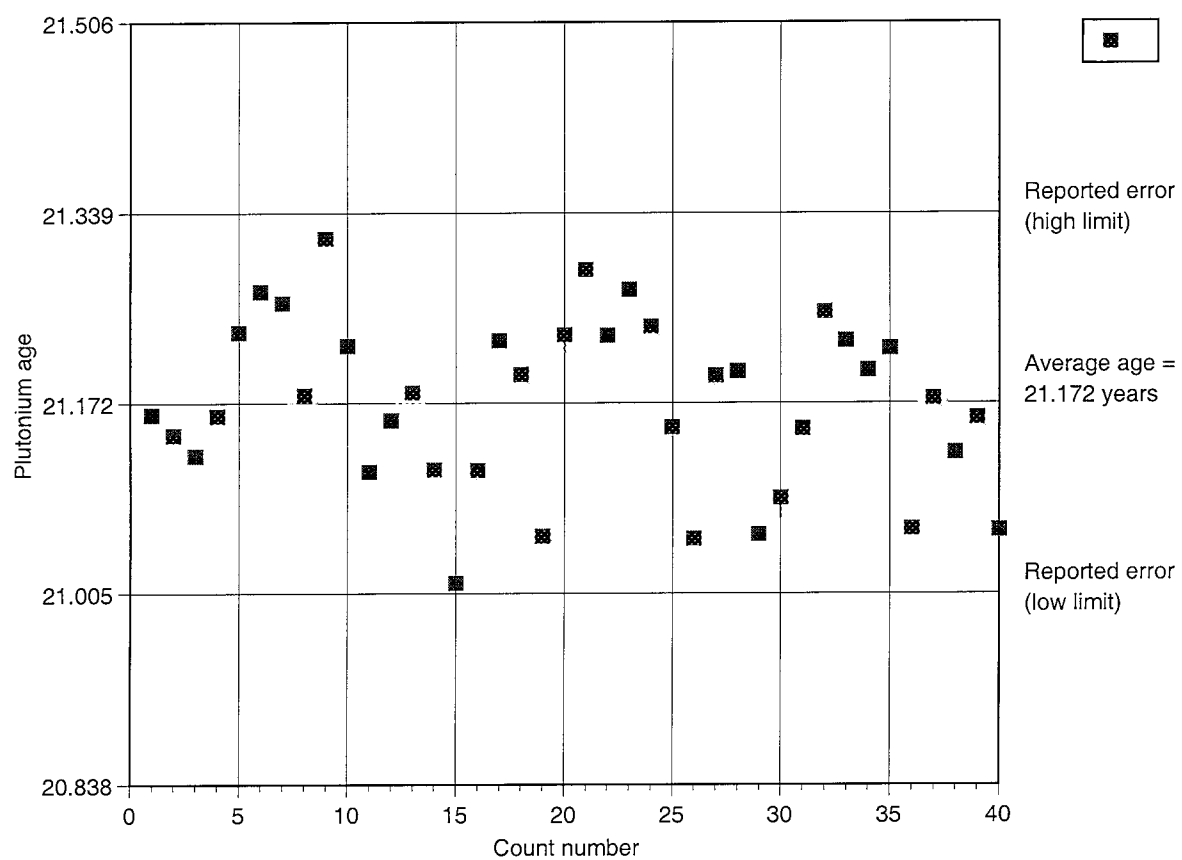


Figure 17. Reported plutonium age precision is better than two sigma